

Measurements of the gate tuned superfluid density in superconducting $\text{LaAlO}_3/\text{SrTiO}_3$

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Abstract

The interface between the insulating oxides LaAlO_3 and SrTiO_3 exhibits a superconducting two-dimensional electron system that can be modulated by a gate voltage. While gating of the conductivity has been probed extensively and gating of the superconducting critical temperature has been demonstrated, the question whether, and if so how, the gate tunes the superfluid density and superconducting order parameter is unanswered. We present local magnetic susceptibility, related to the superfluid density, as a function of temperature, gate voltage and location. We show that the temperature dependence of the superfluid density at different gate voltages collapse to a single curve characteristic of a full superconducting gap. Further, we show that the dipole moments observed in this system are not modulated by the gate voltage.

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Electric field control of conducting channels has allowed great innovation in traditional semiconductor devices [1]. Now heterointerfaces in a new class of materials, the complex oxides, have generated significant interest because of their gate tunable properties. Specifically, the conducting interface formed between the band insulators lanthanum aluminate and TiO_2 terminated 100 strontium titanate (LAO/STO) [2] exhibits many fascinating properties [3] suggesting that an electronic reconstruction triggered by the polar/non-polar interface plays an important role in the inducing the conductivity in the STO [4]. At low temperatures this interface displays two-dimensional superconductivity [5]. Additionally, the high dielectric constant of STO at low temperatures [6] makes applying an electric field with a back gate especially effective to tune the properties of this superconducting state.

Cavaglia *et al.* showed that with increasing gate voltage, V_g , the superconducting critical temperature, T_c , displayed a dome structure and concurrently the normal state resistance monotonically decreased [7]. Later work showed that the electron mobility and carrier density both increased continuously with V_g , with the former dominating the V_g dependence of the conductivity [8]. The evolution of a non-linearity in the Hall resistivity as a function of V_g [8, 9] has been interpreted by Joshua *et al.* as evidence of electrons populating conduction bands with different mobilities [10], implying that the ratio of high and low mobility electrons may be tuned by gating.

Notably, the interface breaks spatial inversion symmetry, opening the possibility for spin orbit coupling to impact the electronic properties of the interface gas. Two groups reported tuning of the Rashba spin orbit coupling (RSOC) inferred from magnetoresistance [11, 12] and measurements of the in-plane critical fields [9]. They found opposite dependencies for tuning the strength of the spin orbit coupling with V_g , making the impact of V_g on the spin orbit coupling unclear, possibly suggesting a peak in the spin orbit coupling.

Moreover, the discovery of magnetic patches coexistent with superconductivity [13–15] and the presence of RSOC originating from the noncentrosymmetric nature of the interface have raised the possibility of an unconventional superconducting pairing mechanism or order parameter [16–18]. However, all previous measurements studying how gating effects the properties of the interface used electronic transport, which gives limited information about the superconducting state. In this Letter, we use local magnetic susceptibility to make the first direct measurements of the superfluid density in LAO/STO and address the question of how the superconducting state evolves with V_g .

Measurements were made on a sample with five unit cells of LAO grown at 800° C and 1.3×10^{-5} mbar oxygen partial pressure on a TiO₂ terminated STO substrate. The growth was followed by a high pressure oxygen anneal, 600° C in 0.4 bar. The sample was silver epoxied to a piece of copper tape, which served as a back gate. V_g was applied between the copper tape and the interface, which was contacted by aluminum wirebonds. Magnetization and susceptibility measurements were made using a scanning SQUID (Superconducting Quantum Interference Device) [19], with a 3 μ m diameter pick-up loop and a concentric field coil for applying a local AC magnetic field. The pick-up loop is sensitive to both the DC static flux and the AC flux resulting from diamagnetic screening currents cancelling the field from the field coil. This setup enables simultaneous measurements of ferromagnetism and superconductivity in the sample [15].

A superconductor will generate screening currents to screen an applied field. The currents extend into a bulk superconductor by the penetration depth, λ . The temperature dependence of λ is a probe of the superconducting state. For a thin superconductor of thickness d , the screening distance is given by the Pearl length $\Lambda = 2\lambda^2/d$ [20]. Using a model by Kogan [1], we extract Λ from measurements of the screening currents as a function of the distance between the sensor and the sample. Λ is related to the superfluid density, $n_s = 2m^*/\mu_0 e^2 \Lambda$, where e is the elementary charge, μ_0 the permeability of free space, and $m^* = 1.46 m_e$ the effective electron mass measured by [22] from Shubnikov de Haas on LAO/STO interfaces. We repeat these measurements at multiple temperatures and gate voltages to map out the superconducting state, Fig. 1. We define T_c as the temperature at which the diamagnetic screening drops below our noise level of $0.01 \Phi_0/\text{\AA}$, corresponding to a minimum detectable n_s of $4 - 14 \times 10^{10} \text{ cm}^{-2}$. The statistical errors were smaller than the systematic errors, outlined in gray in Fig. 2a, from imprecise knowledge of our measurement geometry [23]. The systematic errors are fixed for a single cooldown and represent an overall scaling of n_s which would be the same for every measurement.

T_c vs V_g (Fig. 1a) has a maximum $T_c = 240 \text{ mK}$. In the range of applied V_g superconductivity can only be eliminated on the underdoped side of the dome, and n_s grows monotonically with V_g , with $n_s = 3.0 \times 10^{12} \text{ cm}^{-2}$ at the largest V_g . (Fig. 1b) The carrier density and mobility were measured in a separate cooldown with no backgate. At 2 K the mobility was $1.02 \times 10^3 \text{ cm}^2/\text{Vs}$ and the density was $2.05 \times 10^{13} \text{ cm}^{-2}$, ten times larger than the largest n_s we observed.

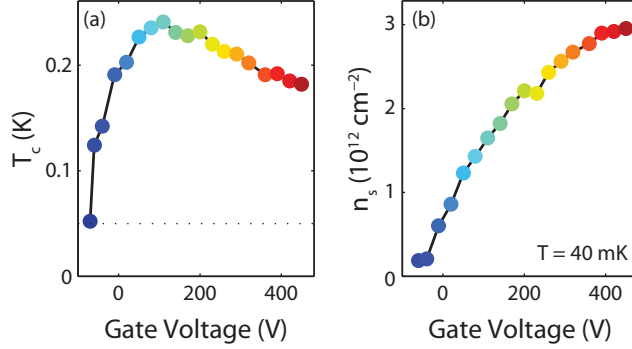


FIG. 1. a) The critical temperature as a function of gate voltage forms a dome. The dashed line represents our lowest measurement temperature. b) The superfluid density at our lowest temperature as a function of gate voltage. The superfluid density increases monotonically throughout the dome. The color scale represents gate voltage and is repeated in Fig. 2.

A small ratio of the superfluid density to the normal density is expected in the dirty limit, in which the elastic scattering time, τ , much shorter than the superconducting gap, Δ_0 ($\hbar/\tau \gg \Delta_0$). \hbar is reduced Plank's constant. Above T_c the normal density of electrons n is given by the optical sum rule $n \propto \int_0^\infty \sigma_1(\omega) d\omega$, where σ_1 is the real part of the conductivity and ω the frequency. For a metal σ is sharply peaked near zero frequency, so scattering moves spectral weight to higher frequencies. Below T_c , a gap opens at $\omega = 2\Delta_0/\hbar$ and the spectral weight within that gap collapses to a delta function at the origin whose amplitude is proportional to n_s [24]. Therefore in the dirty limit, only a fraction of carriers enter the superconducting state, $n_s/n = 2\Delta_0/(\hbar/\tau)$. Using the gate tuned mobilities reported by Bell *et al.*, $100 - 1000 \text{ cm}^2/\text{Vs}$ [8], we expect the ratio n_s/n to be $0.01 - 0.1$, consistent with our measured n_s .

We now look at the temperature dependence of the superfluid density. Fig. 2b plots n_s vs. T for all V_g across the dome. Strikingly, when normalizing the curves they collapse (See Fig. 2c), showing that within our experimental errors there is no change in the superconducting gap structure with electrostatic doping. Furthermore, the collapse is reproducible over multiple positions, sweeps of V_g , and samples [25].

The temperature dependence of the superfluid density is a direct probe of the superconducting order parameter. It can be used to distinguish BCS superconductors from unconventional superconductivity. We fit the normalized curves to a phenomenological BCS model with two parameters Δ and a [2]. Δ scales the superconducting gap $\Delta_0 = \Delta k_B T_c$. a is a

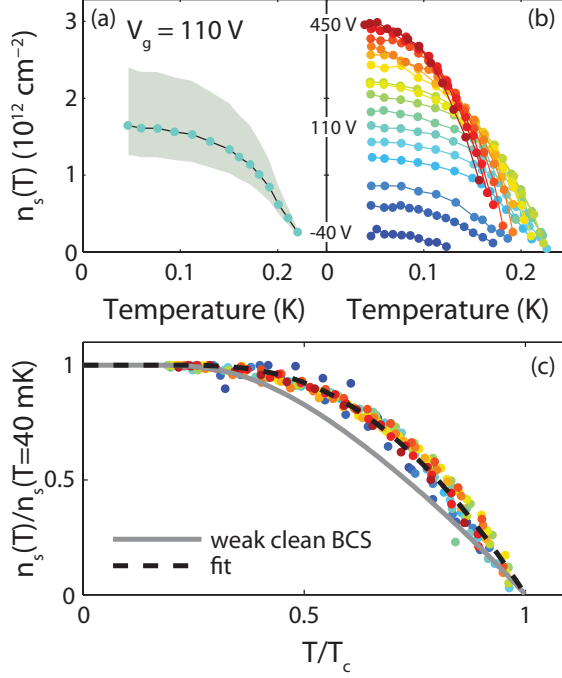


FIG. 2. a) Superfluid density vs. temperature for $V_g = 110\text{V}$, the peak of the superconducting dome. The gray area shows systematic error. b) Superfluid density vs. temperature for every gate voltage. The colors represent the same V_g from Fig. 1. c) Normalized curves from b). The gray line shows the temperature dependence of a weakly interacting clean BCS s-wave superconductor ($\Delta = 1.76$ and $a = 1$). The black dashed line is a fit to the data ($\Delta = 2.2$ and $a = 1.4$).

shape parameter that determines how rapidly the gap opens below T_c , $n_s \propto 1 - (T/T_c)^{2a}$ [27]. $\Delta = 1.76$ and $a = 1$ for an clean s-wave BCS superconductor with weak coupling [2], plotted as the gray line in Fig. 2c. The fit to our data gives $\Delta = 2.2$ and $a = 1.4$. This is consistent with a BCS description with increased coupling or disorder. Both will theoretically increase the gap and the a parameter [28], shifting the curve up and to the right.

The flattening at low temperature indicates fully gapped behavior with a gap that is larger than BCS weak-coupling s-wave. Our lowest measurement temperature is $1/6$ of T_c^{max} , and n_s remains flat (within 3%) up to 35% of T_c . A full gap indicates the absence of low energy quasiparticle excitations, ruling out order parameters with nodes in the Fermi surface. Furthermore, the steep rise of n_s near T_c and the absence of a kink in the functional form rule out most weak coupling two band models [29], because a second smaller gap will slow the onset of superconductivity near T_c . Two gaps of similar size, both larger than the

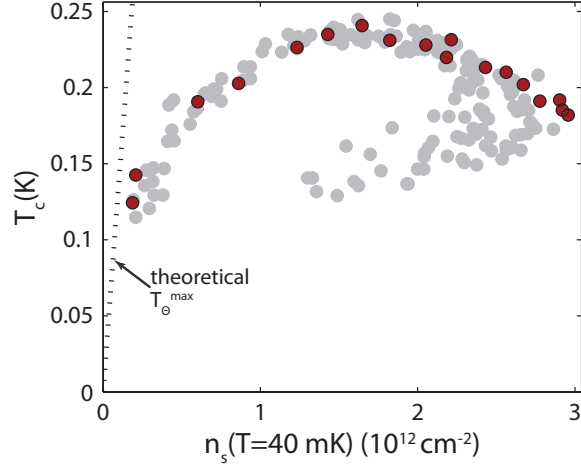


FIG. 3. Critical temperature vs. the superfluid density at lowest temperature ($T \sim 40$ mK). The red points are the data from Figure 1 and the gray dots represent additional data sets. The dotted line is the theoretical phase fluctuation temperature from ref [30], which may be limiting the critical temperature on the underdoped side of the dome. The bimodal distribution on the overdoped side is due to inhomogeneity that locally suppresses n_s in different regions of the sample while the T_c remains the same. See also Fig. 4.

BCS gap or a dominant single large gap with second smaller amplitude gap, could reproduce the data.

The low n_s in the underdoped region may result in suppression of T_c by thermal phase fluctuations. Such fluctuations would result in a linear temperature dependence of n_s in the underdoped region. Following reference [30], we calculate a phase ordering temperature, $T_\theta^{max} = A\hbar^2 n_s(0)/4m^*$, where $A = 0.9$ in two dimensional systems. Fig. 3 shows T_c vs $n_s(40 \text{ mK})$, additionally T_θ^{max} is plotted as a linear function of n_s : the line does not suggest a fit to our data. We have insufficient data at the lowest superfluid densities to make any statement about the functional form of $T_c(n_s)$ in the region where phase fluctuations may be limiting T_c . Nevertheless, the proximity of the phase ordering line to the underdoped data suggests that phase fluctuations may drive the abrupt decrease of T_c .

Given the 2D nature of the superconducting system we expect a BKT transition, where unbinding of vortex anti-vortex pairs suppresses superconductivity and results in a discontinuous jump in n_s near T_c . The jump should occur at finite superfluid density $n_s = 2m^*T_c/\pi\hbar^2$ [31]. For the maximum $T_c = 240 \text{ mK}$ a BKT transition should occur at $5 \times 10^{10} \text{ cm}^{-2}$, which

is too close to our measurement threshold to establish a BKT jump in our n_s vs. T curves.

Are our observations consistent with a simple s -wave order parameter from doped STO [32] or a two gap mixed state induced by symmetry breaking at the interface? Rashba spin orbit coupling (RSOC), induced by the structural inversion asymmetry, is expected to lift the spin degeneracy and split the energy bands [33]. Additionally, RSOC breaks parity and consequently mixes singlet and triplet states resulting in an s -wave component Δ_s mixed with a triplet induced d-vector $\mathbf{d}(\mathbf{k}) = \hat{x}k_y - \hat{y}k_x$ [18, 34]. Mixing results in two gaps, $\Delta = \Delta_s \pm |d_k|$, whose magnitudes depend on the weights of the singlet and triplet components. Varying the relative weights changes the density of states, but always results in two fully gapped Fermi surfaces except for the special case where the s -wave singlet and triplet gaps are the same and accidental line nodes form on one band [18].

Other reports [9, 11] have demonstrated significant tuning of the strength of RSOC with V_g . An open question, of particular importance to testing this two gap picture, is how do the weight of the two components change with V_g . Our results, showing a consistent functional form for n_s vs. T across all V_g , show that the superconducting gap structure does not change with V_g consequently the relative gap weights do not change with V_g . The effect of RSOC on the band structure may depend on the chemical potential which is also tuned by the gate. Therefore the insensitivity of superconductivity to V_g cannot completely rule out a RSOC induced two gap scenario. Yet, our second observation of the fast opening of the gap near T_c and the compatibility of the data with a single gap BCS model limits two gap models. Both gaps must be larger than the BCS s -wave gap to capture both the fast rise and flat low temperature dependence of the data [35].

Finally, disorder may play a role in washing out the triplet component. As stated above, the LAO/STO system is a dirty superconductor, with $\hbar/\tau \gg \Delta$. Disorder averaging has very little impact on the isotropic s -wave component but may eliminate the triplet component.

In short, our data is most consistent with a single gap. We cannot rule out the presence of two gaps, but our observations limit their size and V_g dependence.

Our scanning SQUID system allows two dimensional mapping of superconductivity and magnetism at different V_g . Fig. 4 shows simultaneously imaged susceptometry and magnetometry scans of the same region at 80 mK for four different V_g . The inhomogeneity in the diamagnetic screening is very large in the underdoped region ($V_g = -10$ V) and re-enters

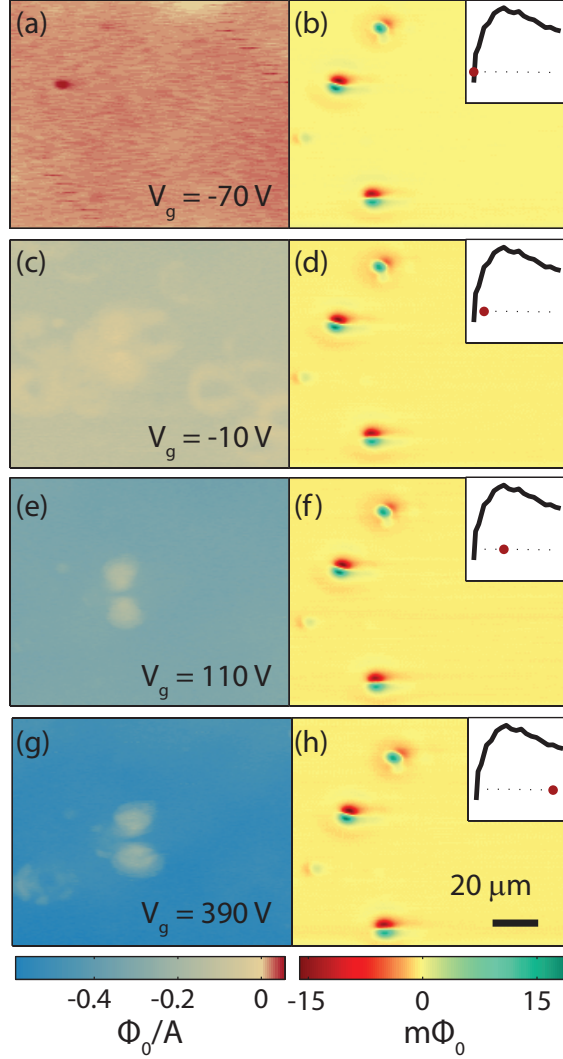


FIG. 4. Susceptometry (left) and magnetometry (right) at 80 mK at different gate voltages. (Inset) Reproduction of the T_c dome from FIG 1 showing the relative location of V_g in each panel. a-b) The sample is no longer superconducting and has a paramagnetic response. Individual ferromagnetic dipoles are also visible in the paramagnetic image. c-d) Superconductivity appears and the landscape is relatively inhomogeneous. e-f) Peak of the superconducting dome, most inhomogeneity disappears. g-h) Excess inhomogeneity returns on the overdoped side of the dome. The ferromagnetic patches do not change with V_g and remain when superconductivity is gone.

the image in the overdoped region ($V_g = 390$ V). The least inhomogeneity is observed at optimal doping, although it does not disappear. In contrast the ferromagnetic patches are insensitive to V_g with a constant magnitude and orientation for all V_g . This behaviour was also observed on 15, 10 and 3.3 uc samples, showing the electron density that is modified

by V_g does not appear to influence the ferromagnetism.

In conclusion, we presented the first measurements of the superfluid density as a function of temperature at multiple gate voltages throughout the superconducting dome in LAO/STO heterostructures. The temperature dependence of n_s is well described by a fully gapped BCS model. Moreover, the normalized n_s vs. T curves collapse to a single functional form indicating there is no change in the gap structure with V_g . Although we cannot rule out a two gap mixed singlet/triplet model, the insensitivity of the superconducting state to V_g and the large slope near T_c limit two gap scenarios. Specifically, both gaps must be larger than the BCS s-wave gap and their relative size cannot change throughout the dome. A future experiment to distinguish between these two scenarios may be to gate the superconductivity in the presence of an in-plane field, which can change the relative magnitude of triplet and singlet gaps. Alternatively, samples in the clean limit may reveal a clearer two gap structure. Additionally, we found that the magnitude and orientation of the ferromagnetic patches that coexist with superconductivity are unchanged by V_g , while at the same time n_s goes from zero to $3.0 \times 10^{12} \text{ cm}^{-2}$. This shows the population of electrons that is modified by the gate is separate from the electrons that contribute to the ferromagnetic order.

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SOM: Gate tuned superfluid density measurements of superconducting $\text{LaAlO}_3/\text{SrTiO}_3$

DISCUSSION OF SYSTEMATIC ERRORS

The accuracy of our superfluid density measurement is dominated by systematic errors which arise from insufficient knowledge of the physical parameters of our SQUID sensor and piezoelectric scanner. Our measurement of the superfluid density, n_s , relies on extracting the Pearl length, Λ , from fits to approach curves.

$$n_s = \frac{2m^*}{\mu_0 e^2 \Lambda} \quad (1)$$

An approach curve measures the diamagnetic susceptibility as a function of the sensor height above the sample. Our SQUID sensor consists of a pair of concentric current carrying wires called the field coil and pick-up loop.

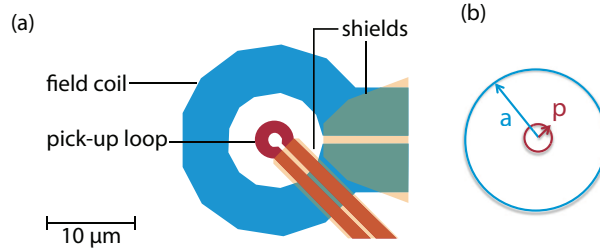


FIG. 5. a) Actual layout of the SQUID field coil, pick-up loop and shields. b) Approximations to the actual layout used by Kogan [1].

We follow a model developed by Kogan which treats the SQUID's field coil as a circular current loop of radius, a [1]. When the loop is brought near a superconducting thin film, the Meissner response of the film detected by the pick-up loop can be expressed as

$$\Phi(h) = \mu_0 \pi a p \int_0^\infty dk \frac{1}{1 + \Lambda k} e^{-2kh} J_1(ka) J_1(kp), \quad (2)$$

where p is the pick-up loop radius, and μ_0 is the magnetic constant. This integral gives a value for the diamagnetic susceptibility, Φ , at a height h above the sample. Six physical parameters enter equation (2): the radius of the pick-up loop p , the radius of the field coil a , the piezo calibration from volts to microns V_c , the distance between the pick-up loop and

the sample when the SQUID makes contact h_0 , the offset of the susceptibility far from the sample Φ_{off} , and a background slope m . We convert the voltage applied to the z-bender, V_z , to a height $h = V_c V_z + h_0$. The susceptibility seen by the SQUID is

$$\Phi_{SQ} = \Phi + \Phi_{off} + mh. \quad (3)$$

Consequently, our fits for Λ depend on the accuracy of our knowledge of the other parameters.

We start with estimates of p and a . We can make accurate measurements of the two radii using an optical microscope; however these wire loops have a finite width and leads that deform their magnetic response with respect to the perfectly circular loops in Kogan's model. Using numerical methods, we calculate the source field using the measured dimensions of our non-ideal coils. We find the non-ideal nature of the field coil and pick-up loop results in a 15% error on the fitted value of Λ . This error works out to few hundred microns on our shortest Λ fits.

We now address the errors associated with our bender constant V_c and height offset h_0 . We don't have accurate calibrations for these parameters, but we do know that these values should be the same for every touchdown curve. We fit hundreds of approach curves using Λ , V_c , and h_0 as free parameters, and assembled histograms of the fitted V_c and h_0 values. From the histograms we were able to extract a best value and variance, σ . We then use the error propagation equation to relate the variances in V_c and h_0 to an error in Λ .

$$\sigma_\Lambda^2 \simeq \sigma_{V_c}^2 \left(\frac{\partial \Lambda}{\partial V_c} \right)^2 + \sigma_{h_0}^2 \left(\frac{\partial \Lambda}{\partial h_0} \right)^2 + \dots + 2\sigma_{V_c h_0}^2 \left(\frac{\partial \Lambda}{\partial V_c} \right) \left(\frac{\partial \Lambda}{\partial h_0} \right) + \dots \quad (4)$$

The propagation equation yielded an error of about 1 mm on our shortest Λ fits. This is a systematic error and is the same for every touchdown curve in the cooldown. It may change the overall calibration for n_s , but it will not change the trends in n_s vs V_g or n_s vs T .

We added the systematic errors from the sensor coils, bender calibration and height offset. The total systematic error is show as the gray outline shown in Fig. 2a of the main text. The error from the bender and offset dominates the error from the non-ideal nature of the pick-up loop and field coil.

DISCUSSION OF PHENOMENOLOGICAL BCS FITS

We compare our normalized plots of superfluid density vs. temperature to a phenomenological BCS model with an isotropic s-wave superconducting gap. The normalized superfluid

density, $n_s/n_s(T=0)$, was given by Prozorov and Giannetta [2]

$$\frac{n_s}{n_s(T=0)} = 1 - \frac{1}{2T} \int_0^\infty \cosh^{-2} \left(\frac{\sqrt{\epsilon^2 + \Delta^2(T)}}{2T} \right) d\epsilon, \quad (5)$$

where T is the temperature and $\Delta(T)$ is the superconducting gap function. The gap can be written [3] as

$$\Delta_0(T) = \Delta_0(0) \tanh \left(\frac{\pi T_c}{\Delta_0(0)} \sqrt{a \left(\frac{T_c}{T} - 1 \right)} \right). \quad (6)$$

$\Delta_0(0)$ is the zero temperature energy gap and a is a shape parameter which determines how fast the gap opens. Near the critical temperature the superfluid density can be approximated as $n_s = 1 - (T/T_c)^{2a}$. For an isotropic s-wave gap $\Delta_0(0) = 1.76k_B T_c$ and $a = 1$. We use equations (5) and (6) to fit our data and find $\Delta_0(0) = 2.2k_B T_c$ and $a = 1.4$. This is the dashed line plotted with the data in Fig. 2c of the main text.

DISCUSSION OF TWO GAPS IN BCS

We can use equations (5) and (6) to generate a phenomenological two-gap expression [4].

$$n_s(T) = p n_{s1}(T) + (1 - p) n_{s2}(T) \quad (7)$$

Fig. 6 shows plots of the superfluid density for two gaps of equal weight ($p = .5$) with different physical parameters. The only combination that can support a superfluid density function that rises faster than BCS near T_c has two gaps that are larger than the BCS gap.

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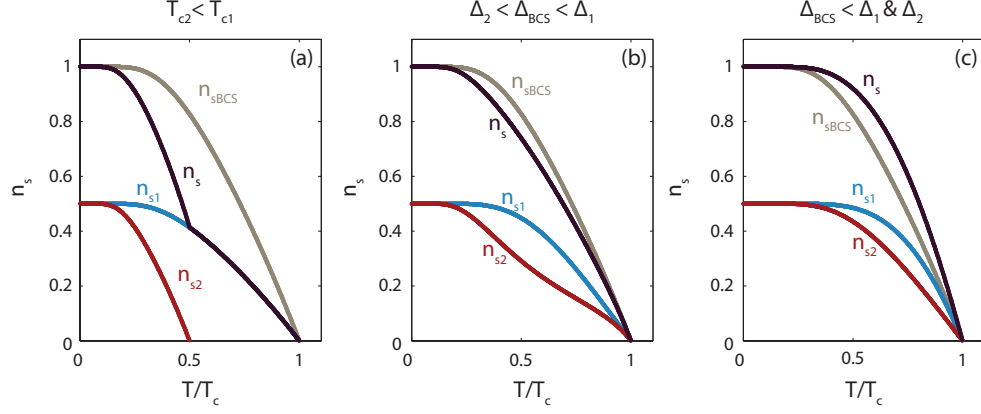


FIG. 6. Comparison of a two-gap superfluid density n_s with a single gap BCS superfluid density $n_{s\text{BCS}}$. In all three plots $n_{s1} = pn_{s1}$ and $n_{s2} = pn_{s2}$ with a) Plots of two gaps with $\Delta_1 = \Delta_2 = 1.76$ and $a_1 = a_2 = 1$ but two different critical temperatures. b) Plots of two gaps with $\Delta_1 = 2.2$, $\Delta_2 = 1.1$, and $a_1 = a_2 = 1$. c) Plots of two gaps with $\Delta_1 = 3$, $\Delta_2 = 2$, $a_1 = 1.8$ and $a_2 = 1$. Only in c) where both gaps are larger than $\Delta_{\text{BCS}} = 1.76$ can we generate a total superfluid density that opens faster than BCS.